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Research Article

Essential oil composition of four *Teucrium* L. taxa from Turkey, their chemotaxonomy and potential usefulness

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ABSTRACT

In this study, the essential oil components of aerial parts of four *Teucrium* (*T. chamaedrys* L. subsp. *tauricolum* Rech.f., *T. parviflorum* Schreb., *T. polium* L. and *T. orientale* L. var. *orientale*) taxa were investigated by GC and GC-MS. Thirty three, thirty two, thirty five and twenty eight components were detected representing 93.08%, 89.10%, 92.20% and 89.14% of the oil, respectively. Germacrene D (20.17%), caryophyllene oxide (17.05%), β-pinene (13.45%); α-pinene (23.62%), β-caryophyllene (15.90%), β-pinene (14.65%); β-pinene (20.38%), *p*-cymene (16.54%), α-pinene (16.51%), caryophyllene oxide (5.92%) and germacrene D (19.45%), β-caryophyllene (18.95%), β-pinene (12.56%) were determined the main compounds of *T. chamaedrys* subsp. *tauricolum*; *T. parviflorum*; *T. polium* and *T. orientale* var. *orientale*, respectively. The results were discussed in means of chemotaxonomy, natural products and potential usable of these plants.

Key words: *Teucrium*, essential oil, chemotaxonomy, natural product.

INTRODUCTION

The genus *Teucrium* (Lamiaceae) comprises more than 300 taxa; almost 50 are known in Europe and distributed chiefly in the Mediterranean basin[1]. In the Flora of Turkey *Teucrium* is represented by 42 taxa[2-5]. Perennial herbs or small shrubs and divided eight sections (Teucrium, Scordium, Chamaedrys, Polium, Isotriodon, Stachybotrys, Scorodonia, Spinularia). *T. orientale* var. *orientale* and *T. parviflorum* are in the Teucrium section; *T. chamaedrys* subsp. *tauricolum* is in the Chamaedrys section; *T. polium* is in the Polium section. *T. chamaedrys* subsp. *tauricolum* is an endemic plant in Flora of Turkey[2].

Micromorphological characters, conspicuously trichomes, are one of the most useful taxonomic properties in *Teucrium* genus. Their absence or presence and also their typology have a important role in classification of this genus. Several studies are present on *Teucrium* morphology and micromorphology and has useful information about trichome distribution on the calyx, corolla, leaves, and nutlets of species of the Mediterranean area[6,7].

Medicinal and aromatic plants are believed to be an important source of new chemical substances with potential therapeutic activities[8]. This genus is one of the richest resources of diterpenes, with a neoclerodane skeleton: more than 220 diterpenes have been described up to now, and many of these diterpenes are especially interesting owing to their ecological role as antifeedants opposite different species of insects and for their role in the medicinal features of the plants[9]. *Teucrium* taxa have been used as medicinal plants for more than 2000 years, some of them are still used in folk medicine; for their biological properties as tonic, antispasmodic, antipyretic, antiseptic[10] and hypoglycaemic, hypolipidemic, hepatoprotective, antipyretic, anti-inflammatory, antiulcer, antitumor, antibacterial activities[11,12]. Moreover, *Teucrium* taxa have been used in the food industry to uses chemical preservatives, to prohibit the growth of food spoiling microbes[13]. The importance of *Teucrium* and Lamiaceae family paterrns in food industries lies also on the fact that many *Teucrium* species have antioxidant, antimicrobial and antifungal activities, suplly them useful as natural preservative ingredients[14-16].

The objective of the present study was (a) to examine the chemical composition of the essential oil of *T. chamaedrys* subsp. *tauricolum*, *T. parviflorum*, *T. polium* and *T. orientale* var. *orientale* by GC and GC-MS, growing wild in the eastern part of Turkey (b) to evaluate the medicinal, agricultural purpose, natural products, renewable resources and chemotaxonomy of the studied samples.

MATERIALS AND METHODS

PLANT MATERIAL

Plant samples were collected from their natural habitats. *T. orientale* var. *orientale* was collected from west of Dikme village (Bingöl) steppe, on 20.06.2013, at an altitude of 1400-1500 m., by O. Kilic (4789). *T. parviflorum* and *T. polium* were collected from vicinity of Dikme upland (Bingöl) slopes, on 30.06.2013, at an altitude of 1400-1500 m., by O. Kilic (5153-5155). *T. chamaedrys* subsp. *tauricolum* was collected from north of Dikme upland (Bingöl), edge of *Quercus* forest, on 30.06.2013, at an altitude of 1450-1500 m., by O. Kilic (5161). All plant samples were identified by Kilic with Flora of Turkey and East Aegean Islands (2). The voucher specimens have been deposited at the Herbarium of Biology Department (BIN), Bingol University.

ISOLATION OF THE ESSENTIAL OIL

Air-dried aerial parts of the *Teucrium* taxa were subjected to hydrodistillation using a Clevenger-type apparatus for 3 h.

GAS CHROMATOGRAPHIC (GC) ANALYSIS

The essential oil of studied species was analyzed using HP 6890 GC equipped with and FID detector and an HP-5 MS column (30m × 0.25mm i.d., film tickness 0.25μm) capillary column was used. The column and analysis conditions were the same as in GC-MS. The percentage composition of the essential oils was computed from GC-FID peak areas without correction factors.

GAS CHROMATOGRAPHY-MASS SPECTROMETRY (GC-MS) ANALYSIS

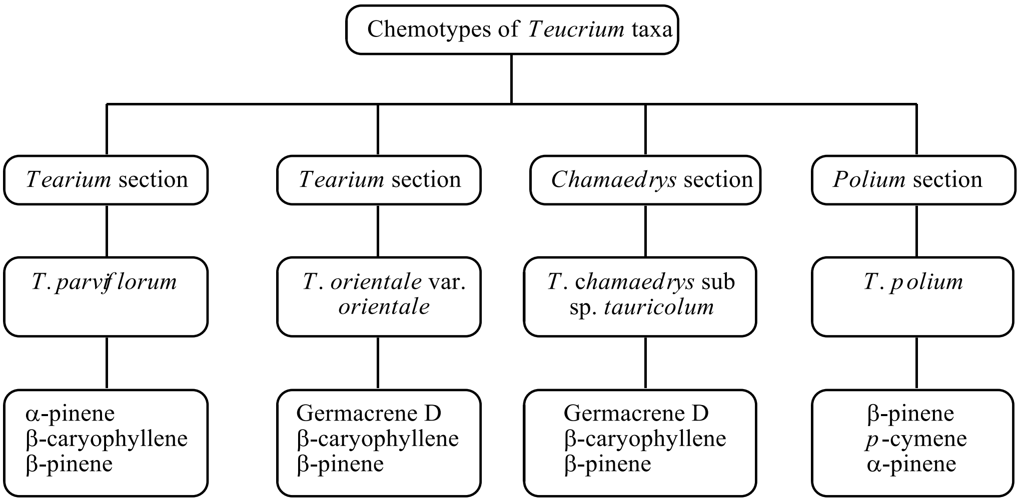
The essential oils of four *Teucrium* taxa were analyzed by GC, GC-MS, using a Hewlett Packard system. HP-Agilent 5973 N GC-MS system with 6890 GC in Plant Products and Biotechnology Research Laboratory (BUBAL) in Firat University. HP-5 MS column (30m × 0.25mm i.d., film tickness (0.25μm) was used with helium as the carrier gas. Injector temperature was 250ºC, split flow was 1ml/min. The GC oven temperature was kept at 70ºC for 2 min and programmed to 150ºC at a rate of 10ºC/min and then kept constant at 150ºC for 15 min to 240ºC at a rate of 5ºC/min. Alkanes were used as reference points in the calculation of relative retention indices (RRI). MS were taken at 70eV and a mass range of 35-425. Component identification was carried out using spectrometric electronic libraries (WILEY, NIST). The identified compounds are showed in Table 1 and the chemotypes of *Teucrium* samples are seen in Figure 1.

RESULTS AND DISCUSSION

In this study, *T. chamaedrys* subsp. *tauricolum*, *T. parviflorum*, *T. polium* and *T. orientale* var. *orientale* were examined by GC and GC-MS. Germacrene D (20.17%), caryophyllene oxide (17.05%) and β-pinene (13.45%) were the main compounds of *T. chamaedrys* subsp. *tauricolum*; α-pinene (23.62%), β-caryophyllene (15.90%) and caryophyllene oxide (14.65%) were reported the major constituents of *T. parviflorum*; β-pinene (20.38%), *p*-cymene (16.54%), α-pinene (16.51%) and caryophyllene oxide (5.92%) were determined the main constituents of *T. polium*; germacrene D (19.45%), β-caryophyllene (18.95%) and β-pinene (12.56%) were determined the main compounds of *T. orientale* var. *orientale*.

The chemical composition of *T. libanitis* Schreb. and *T. turredanum* Losa & Rivas Goday collected at different localities were analysed by GC and GC-MS. α-pinene (9.9-21.2%) and *δ*-cadinene (5.3-9.7%) were the predominant compounds of *T. libanitis*, while that of *T. turredanum* had a higher content of β-caryophyllene (15.6-32.6%), α-humulene (4.7-10.1%) and β-bisabolol (6.4-8.3%)[17]. In our study, α-pinene (23.62-16.51%) was among the predominant compound of *T. parviflorum* and *T. polium* respectively, on the other hand α-pinene was not predominant constituent in the oil of *T. chamaedrys* subsp. *tauricolum* and *T. orientale* var. *orientale* (Table 1).

Fig. 1: Chemotypes of studied East Anatolian (Bingol) *Teucrim* taxa



Germacrene D (20.17% - 19.45%) was found to be the major constituent of *T. chamaedrys* subsp. *tauricolum* and *T. orientale* var. *orientale* oils respectively; this compound also has been reported in the *T. pestalozzae* Boiss. (13.8%), *T. sandrasicum* O. Schwarz (27.9%), *T. montanum* L. (5.8%)[18] and *T. antitauricum* T.Ekim oils[19]; whereas germacrene D was found to be low amount in the essential oils of *T. parviflorum* (4.04%) and *T. polium* (3.98%) (Table 1). β-caryophyllene was the most abundant component in *T. orientale* L. var. *puberulens* T.Ekim (21.7%), *T. chamaedrys* L. subsp. *lydium* O.Schwarz (19.7%)[20] and in *T. antitauricum* (27.6%)[19]; similarly in this research β-caryophyllene contained high concentrations of *T. parviflorum* (15.90%) and *T. orientale* var. *orientale* (18.95%) (Table 1).

In another study with *Teucrium scordium* L. the major constituents of the oil were β-caryophyllene (22.8%), (*E*)-β-farnesene (10.4%), caryophyllene oxide (8.6%), 1,8-cineole (6.1%) and β-eudesmol (5.1%)[21]; like this study, caryophyllene oxide (17.05%, 14.65%) was determined the major constituents of the *T. chamaedrys* subsp. *tauricolum* and *T. parviflorum*, respectively (Table 1). In the essential oil analysis of the four *Teucrium* taxa in here showed some similarities with the Morteza-Semnani et al., (2005) study; they determined that, the major constituents of studied plant oil were germacrene D (16.5%), (Z) β-farnesene (12.2%), β-caryophyllene (10.5%) and α-pinene (9.1%) like in our samples with different quantity; β-farnesene was not detected or detected only low amount in our study (Table 1). Kovacevic & Lakusic (2001), determined 32 constituents in the leaves of *T. chamaedrys* collected from Serbia and Montenegro, they also determined that β-caryophyllene (26.9%) and germacrene D (22.8%) to be the major constituents in this species[22]. When we compared the essential oil composition of *T. chamaedrys* subsp. *tauricolum*, *T. parviflorum*, *T. polium* and *T. orientale* var. *orientale* sample from Turkey; there was a contrast in the quantity of these two components as not determined among the main compounds of *T. polium* (Table 1). The analysis of the essential oil of *T. multicaule* was characterized by a higher content of caryophyllene oxide (32.1%) and thymol (14.6%); β-caryophyllene (19.6%) and germacrene D (12.3%) were in *T. parviflorum*[23]. In addition cited studies, there are some researchs about chemotaxonomy and potential usefulness of Lamiaceae family[24-26].

We can say that the differences in the quality or quantity of the composition of essential oils may be due to genetical, drying conditions, differing chemotypes, method of distillation and / or extraction and geographic or climatic factors. In conclusion from the research, it can be said that the essential oil of *T. chamaedrys* subsp. *tauricolum* has germacrene D/caryophyllene oxide; *T. parviflorum* has α-pinene/β-caryophyllene; *T. polium* has β-pinene/*p*-cymene and *T. orientale* var. *orientale* has germacrene D/β-caryophyllene types from their essential oils. In addition, *T. orientale* var. *orientale* and *T. parviflorum* are in the Teucrium section; so the results of this study overlaps with the morphological classification, except for germacrene D and α-pinene percentages. These results have also ecological and economic significance for utilization of the species in the medicinal, agricultural, cosmetic and chemical industries.

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Table 1: Essential of composition of studied *Teucrium* taxa

Compound	RRI*	*T. chamaedrys* subsp. *tauricolum*	*T. parviflorum*	*T. polium*	*T. orientale* var. *orientale*
2-Hexenal	865	0.12	-	0.33	-
α-pinene	930	5.52	23.62	16.51	4.52
β-pinene	955	13.45	14.65	20.38	12.56
β-myrcene	985	-	1.03	-	1.53
p-cymene	1010	2.05	4.21	16.54	3.54
1,8-cineole	1022	2.65	-	2.13	-
Camphene	1032	-	1.14	-	0.21
Sabinene	1048	4.06	2.96	3.47	-
Linalool	1075	-	-	1.49	5.43
Nonanal	1082	0.14	0.21	-	-
trans-sabinenehydrate	1115	0.30	-	0.14	1.03
α-terpinolene	1128	-	0.13	1.04	-
Bornyl acetate	1136	1.04	0.71	-	0.45
Trans-chrysanthenol	1160	0.21	-	0.14	1.01
α-terpineol	1174	-	1.24	0.31	-
Myrtenol	1190	0.21	0.11	-	0.10
3-cyclohexen-1-ol	1215	0.32	-	0.24	0.14
2-Cyclohexen-1-one	1248	-	0.14	0.21	-
Thymol	1292	0.74	0.85	1.02	0.74
Carvacrol	1305	0.45	1.05	-	1.32
Propanocacid	1342	0.25	-	0.12	-
α-copaene	1360	1.21	0.52	1.05	-
β-cubebene	1402	-	0.34	-	2.14
Aromadendrene	1415	1.02	-	0.54	-
γ-decalactone	1420	1.03	0.25	-	0.41
β-caryophyllene	1428	4.85	15.90	3.85	18.95
Ledene	1438	-	0.14	0.09	-
Germacrene D	1448	20.17	4.04	3.98	19.45
(*E*)-β-Farnesene	1452	3.54	-	4.41	3.54
α-humulene	1458	2.64	5.58	3.75	-
Spathulenol	1465	-	-	0.24	4.12
Elemol	1448	0.21	0.52	-	0.16
β-bisabolol	1461	0.45	1.23	2.08	-
β-bisabolene	1470	1.50	0.85	1.12	0.45
β-sesquiphellandrene	1485	0.14	-	0.08	0.14
α-selinene	1492	0.89	-	-	0.21
Caryophyllene oxide	1505	17.05	4.10	5.92	2.01
β-bisabolene	1512	-	1.24	0.12	-
γ-cadinene	1513	5.24	-	3.14	-
α-cadinol	1540	-	9.34	-	4.00
Azulene	1605	0.05	-	0.74	0.32
2-pentadecanone	1635	-	0.54	-	-
Hexadecanoicacid	1695	0.24	0.12	0.25	0.41
Phytol	1785	0.14	-	1.01	-
Cis-calamenene	1796	-	0.14	0.50	-
Tricosane	1895	0.20	0.05	0.12	0.25
Ericosane	1936	-	0.15	0.14	-
	Total	93.08	89.10	92.20	89.14

*RRI: Relative Retention Index.

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